

"SLEEPING REACTOR" IRRADIATIONS: A NOVEL USE OF HFIR IN THE DETERMINATION OF ELEMENTS WITH SHORT-LIVED ACTIVATION PRODUCTS. E.A. Jerde¹, D.C., Glasgow², G.A.Snyder³, and L.A. Taylor³, ¹Oak Ridge Research Institute, Oak Ridge, TN 37830 (eoa@ornl.gov); ²Chemical and Analytical Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831; ³Planetary Sciences Institute, Univ. of Tennessee, Knoxville, TN 37996

Introduction

Neutron activation analysis (NAA) is a reliable, sensitive technique for the determination of elemental concentrations that has been widely applied in the study of lunar and meteoritic samples. By varying irradiation times, decay periods, and counting times, a wide range of both major and trace elements can be determined, often down to the part-per-billion level (for detailed discussions of NAA, see [1,2]).

At the High Flux Isotope Reactor (HFIR) at the Oak Ridge National Laboratory, the irradiation system (pneumatic tube 1 or PT-1) has a thermal neutron flux (ϕ) of $\sim 4 \times 10^{14}$ n cm⁻² s⁻¹. This high flux permits the detection of most elements with irradiations as short as 60 seconds. In typical mixed matrices such as rocks or soils, an analysis is often performed through a pair of irradiations. The first is short, 6 or 7 seconds, followed by two counts over the next three days. This permits the determination of elements with half-lives less than 24 hours. A second irradiation is then made, generally 60 seconds or so, followed by a decay period of 6 or 7 days. This induces sufficient activity in the longer-lived species (half-lives >1 day) that detection is possible in counts made over the next month or so. This double irradiation allows the detection of elements of widely varying half-lives, without the accompanying high radioactivity encountered early with the single irradiation scheme.

Problems associated with "ultra-short" half-lives

As mentioned above, elements with short-lived activation products are obtained through very short irradiation times and counts. Due to uncertainties in the timing of the pneumatic system (+/- ~0.5 second), irradiations of less than 6 seconds or so are less reliable. The 6- or 7-second irradiations are quite acceptable for detection of elements with half-lives of as little as 30 minutes. However, geologically important elements such as Al, Mg, Ti, and V have half-lives of only a few minutes. In more typical research reactors, where the neutron flux is perhaps two orders of magnitude lower than at HFIR, irradiations of a minute or two, followed by immediate counting, suffice to determine these elements. At HFIR, these can occasionally be determined with irradiation times ~6 seconds, but the requirement of immediate counting leads to increased exposure to the high activity produced by irradiation in the extremely high flux. As a result, the determination of these "ultra-short" lived species in mixed matrices has not generally been made at HFIR.

The "Sleeping Reactor" solution

Recently, we have found that very short-lived activation products can be made and detected relatively easily and safely during the period after reactor shutdown (SCRAM), but before the spent fuel elements are removed from the HFIR pressure vessel. During this 24-36 hour period which we have dubbed the "sleeping reactor", neutrons are formed in the beryllium reflector surrounding the core by the reaction $^9\text{Be}(\gamma, n)^8\text{Be}$, with the γ -rays coming principally from the spent fuel

element. Upon reactor SCRAM, the flux drops from $\sim 4 \times 10^{14}$ (the typical full-power flux) to $\sim 1 \times 10^{10}$ n cm⁻² s⁻¹ within one hour, mainly due to the cessation of fission-produced neutrons. By the time the fuel elements are being removed (generally 24 hours after SCRAM) the flux has dropped to $\sim 6 \times 10^8$ (Fig. 1). Neutron fluxes in the 10^8 - 10^{10} range can be utilized for the determination of short-lived elements such as Al, Ti, Mg, V, and Ca.

This source of neutrons at HFIR has long been known, but has never been utilized. In the sleeping reactor, we know that the flux is NOT constant, precluding the use of comparative analysis. Therefore, in order to perform an analysis using the sleeping reactor, it must be shown that we have a way of knowing the flux at any given time after SCRAM.

The HFIR fuel elements are all identical, so it can be expected that after shutdown the neutron flux behavior should be the same from cycle to cycle. During the tests run so far, the flux has behaved in an identical fashion after many cycles (Fig. 1 includes 5 cycles). This is the result of two factors. First, the relative abundances of the various fission products building up in the fuel is always the same. Therefore the flux decay curve should have the same shape from cycle to cycle. Second, the principal early decay of the fission products has a "half-life" on the order of a day or two. Thus, the 25-day reactor cycle reaches saturation for these elements.

Initial examinations show that for materials of geologic interest, irradiations of 300 seconds provide adequate counting statistics for the elements in question, yet do not produce any significant residual (i.e., long-lived) activity in the samples. This is the primary benefit; previous work in the determination of short-lived elements, done while the reactor was at full power, created a great deal of activity, adding significantly to the radiation dose received by the experimenter.

Results

The curve in Fig. 1 allows the flux to be calculated with a greater than 99% certainty after SCRAM. A group of standard materials were irradiated as checks to verify our methods. Using the flux as represented by the curve in Fig. 1, the reference materials NIST 2711, USGS SCo-1, and USGS BHVO-1, yielded the results shown in Table 1 below. These values are in good agreement with the accepted values (in parentheses) over an entire day of irradiations, and an order of magnitude change in flux. No other activity of any consequence is created in the analysis of these; a major advantage.

In addition, we have analyzed six Apollo 11 samples to assess the ability to determine these elements in small aliquots of material. The results of these analyses are shown in Table 2. It can be seen that the sleeping reactor method is feasible even for samples well under 100 mg. The missing data in the later samples is due to the lower flux at the time of later irradiation. Obviously, for small samples, it is advantageous

to irradiate them relatively soon after SCRAM. However, due to the rapid reduction in flux during the first 3 hours or so after SCRAM, there is a greater uncertainty in the flux curve. Therefore, practical use of this technique should only begin 3 hours or so after SCRAM.

As a final note, some residual neutron activity exists even after the fuel element removal, due to radioactivity in the europium control plates and the antimony sources used in

reactor startup. Although the flux is low (the order of 10^8 n cm^{-2} s^{-1}) it also seems to act in a predictable fashion. This may be suitable for uses where liquids need to be irradiated, such as medical experiments.

References: [1] Ehmann, W.D. and Vance, D.E. (1991) Radiochemistry and nuclear methods of analysis. Volume 116 in Chemical Analysis: A series of monographs on analytical chemistry and its applications (J.D. Winefordner, Series editor), Wiley, New York. [2] Parry, S.J. (1991) Activation spectrometry in chemical analysis. Volume 119 in Chemical Analysis: A series of monographs on analytical chemistry and its applications (J.D. Winefordner, Series editor), Wiley, New York

Table 1. Results of sleeping reactor irradiations of standard materials.

Sample	T _{SCRAM} (min)	ϕ (n cm ⁻² s ⁻¹)	Al (%)	Ti (%)	Mg (%)	V (ppm)
BHVO-1	319.8	2.60e9	7.34±.11	1.66±.12	5.36±.36	349±16
	472.1	1.77e9	7.23±.10	1.77±.14	4.14±.41	354±16
	530.4	1.58e9	7.15±.08	1.79±.14	4.61±.46	345±15
	1212.2	6.93e8	7.46±.06	1.68±.20	4.62±.5	329±18
	1327.6	6.33e8	7.37±.07	1.60±.20		337±18
Average	-----		7.31±.12 (7.30)	1.70±.08 (1.62)	4.75±.21 (4.36)	343±10 (317)
NIST 2711	1480.6	5.68e8	6.35±.20 (6.5)	0.46±.2 (0.3)		85±24 (82)
SCo-1	1580.5	5.32e8	14.04±.4 (13.7)	0.80±.37 (0.63)	2.41±1.2 (2.7)	116±28 (131)

Table 2. Results of sleeping reactor irradiation of six Apollo 11 basalts.

	mass (mg)	Al ₂ O ₃ (%)	MgO (%)	TiO ₂ (%)	V (ppm)	CaO (%)
10085,1268	48.7	10.15±.26	12.21±2.2	10.94±.95	168±25	
10085,1269	63	12.18±.38	9.67±4.83	11.40±1.18	104±36	14.4±6.4
10085,1271	41.3	10.27±.41	6.21±6.03	11.78±1.47		
10085,1276	42.2	11.49±.46		9.86±1.47		
10085,1280	36.2	10.30±.46		13.28±1.71		
10085,1283	44.6	11.40±.47		11.65±1.53		

